

Construction of One-dimensional Humidifier Model in Fuel Cell System

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ABSTRACT

The phenomena inside a porous hollow fiber membrane humidifier were analyzed and a one-dimensional humidifier model that can estimate the water transfer rate was constructed to enable control of the water content in the fuel cell stack of a fuel cell system to the appropriate range. The calculation accuracy of the humidifier model was enhanced by investigating and considering the impact of the dry-up phenomenon that may lower the water transfer rate due to partial drying of the porous membrane. In addition, verification was performed by comparing the calculation results of the humidifier model with the results of humidifier evaluation tests and measurements of the water transfer rate in bench operation of the fuel cell system. The estimation accuracy of the humidifier model was confirmed to satisfy the target accuracy of $\pm 15\%$ or less that is needed for use in performance design and control optimization of fuel cell systems.

1. Introduction

Polymer electrolyte fuel cells are power generation devices with a low environmental load, and are attracting attention as a next-generation power source.

To help ensure the power generation performance of polymer electrolyte fuel cells, it is necessary to maintain the thickness of the Polymer Electrolyte Membrane (PEM) that functions to conduct hydrogen ions and to physically separate oxygen and hydrogen. OH radicals and hydrogen radicals generated as by-products during power generation chemically cleave the polymer chains of the PEM, and the cleaved polymer chains are exhausted out of the cell, which may reduce the membrane thickness. As a result, the amount of unreacted gas that permeates between the cathode and anode via the PEM increases and more hydrogen is consumed, which affects fuel efficiency performance. Insufficient water content in the PEM may impede radical transfer, resulting in higher concentrations and promoting the reduction in PEM thickness. This is also known to lower the chemical stability of the PEM, which accelerates aging⁽¹⁾⁻⁽³⁾.

Many fuel cell systems adopt humidifiers to humidify the air supplied to the fuel cell stack in order to provide the PEM water content (Fig. 1). High-humidity air (hereinafter defined as WET gas) is exhausted from the fuel cell stack and flows into the humidifier installed in the air supply system. The air supplied to the fuel cell stack (hereinafter

defined as DRY gas) is humidified by the water vapor contained in the WET gas.

However, if the fuel cell stack has an excessive water content, the supply of hydrogen to the anode is impeded, the power generation efficiency drops, and aging is accelerated. It is important to control the relative humidity (RH) of the DRY gas in order to adjust the water content in the fuel cell stack to the appropriate range. This calls for estimation of the water transfer rate due to the humidifier, and multiple studies have been conducted to elucidate the humidification performance of humidifiers applied to fuel cell systems through experiments and modeling^{(4),(5)}.

Honda has adopted a porous hollow fiber membrane humidifier in its fuel cell system that can achieve both humidification performance and differential pressure strength.

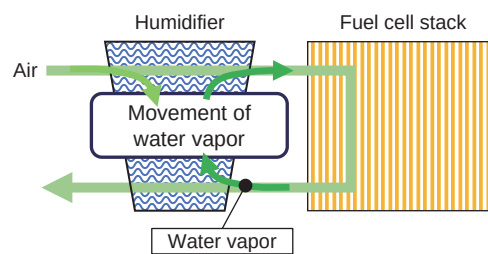


Fig. 1 Humidification system

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2. Development Goals

Porous membranes are characterized by a drop in humidification performance under conditions with low WET gas RH. It is necessary to accurately understand the phenomenon of this drop in humidification performance in order to properly humidify the DRY gas, but to date there is insufficient research on the characteristics of porous membrane humidifiers. This study referred to this phenomenon as the dry-up phenomenon, and analyzed the factors by which it may occur.

A one-dimensional humidifier model that estimates the RH of the air supplied to the fuel cell stack from the operating conditions was constructed for use in performance design and control optimization of fuel cell systems. This humidifier model reproduces the effects of the dry-up phenomenon, and enhances the estimation accuracy under conditions with low WET gas RH. The target accuracy of the humidifier model was set to $\pm 15\%$ or less, considering variance factors in the fuel cell system and taking into account the model estimation error needed to help prevent PEM aging and flooding.

3. Construction of Humidifier Model

3.1. Mechanism of Humidification

This section describes the method by which the humidifier humidifies the fuel cell system. The humidifier functions to self-recover the generated water from the WET gas and to provide the DRY gas with the appropriate humidity needed for power generation. In the fuel cell system, the humidifier and the fuel cell stack form a circulatory system as shown in Fig. 2. Here, water circulates in a cycle in which (I) water vapor transfers due to the partial pressure difference between the WET gas and DRY gas, (II) the water vapor is exhausted from the fuel cell stack along with the water generated by power generation (part of the generated water permeates to the anode), and (III) the WET gas flows into the humidifier. Therefore, the water transfer rate due to the humidifier N_{wtr} [mol/s] is expressed in terms of the amount of water generated by the fuel cell stack N_{gen} [mol/s], the water content in the WET gas N_{wet} [mol/s], and the water transfer rate to the

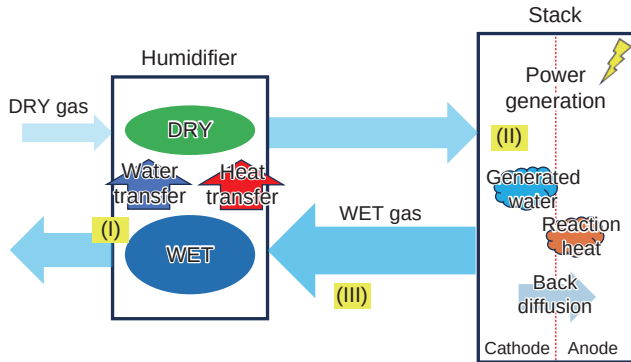


Fig. 2 Water circulation in fuel cell system

anode $N_{backdiff}$ [mol/s], as shown by the relationship in Eq. (1).

$$N_{wtr} = N_{gen} - N_{wet} - N_{backdiff} \quad (1)$$

Heat exchange takes place inside the humidifier between the WET gas, which has risen in temperature due to the reaction heat of the fuel cell stack, and the DRY gas, and this change in the gas temperature influences water transfer.

The humidifier consists of a humidifier module and heads that form flow channels, as shown in Fig. 3. The inside of the humidifier module is filled with thousands of hollow fibers, and DRY gas flows inside and WET gas flows outside the hollow fibers. Figure 4(a) shows a magnified view of the hollow fiber membrane cross section. There are pores on the order of several nm through which water is exchanged. The diameter of the hollow fiber membrane pores becomes smaller from the outer surface (WET gas side) shown in Fig. 4(b) toward the inner surface (DRY gas side) shown in Fig. 4(c)⁽⁶⁾.

The hollow fiber membrane has a cylindrical structure that is advantageous for obtaining high differential pressure strength without additional support structures as compared to a flat membrane. In addition, whereas other exchange

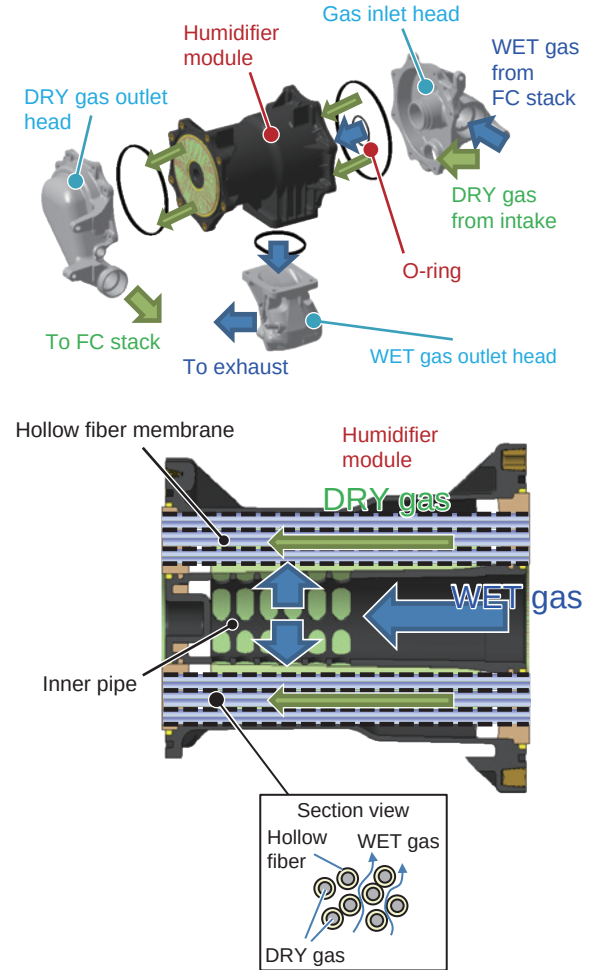
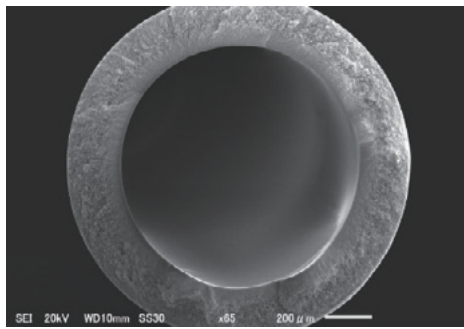


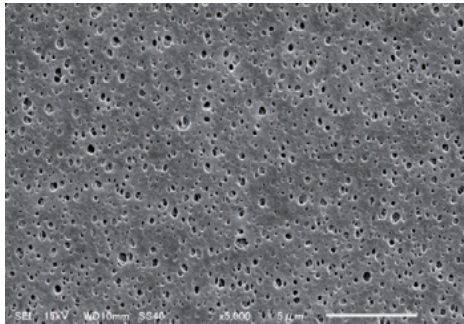
Fig. 3 Structure of humidifier module

membranes such as proton exchange membranes and dissolution diffusion membranes require thinning to increase the water vapor permeability, in porous membranes, the pore diameter acts instead of the thickness. This means that both differential pressure strength and humidification performance can be achieved by providing membrane thickness, which makes porous membranes suitable for use in fuel cell systems.

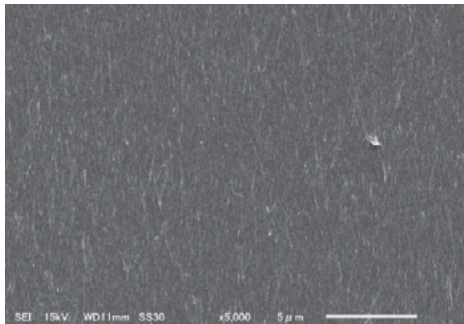
Next, the mechanism of water transfer in the porous membrane is explained⁽⁷⁾. Figure 5 shows the mechanism of water transfer by the pores. The water in the WET gas is attracted to the membrane surface by the adsorption phenomenon, and that water then moves toward the DRY gas by capillary transfer inside the pores. The water that passed through the pores enters the DRY gas by evaporative diffusion due to the sudden drop in pressure. This continuous process is the mechanism of humidification.



(a) Membrane cross section 65X magnification



(b) Outer surface 5000X magnification



(c) Inner surface 5000X magnification

Fig. 4 Magnified views of hollow fiber membrane

3.2. Overview of One-dimensional Humidifier Model

This section describes an overview of the one-dimensional humidifier model for accurately estimating the water transfer rate by the humidifier. Figure 6 shows the flowchart of the one-dimensional humidifier model.

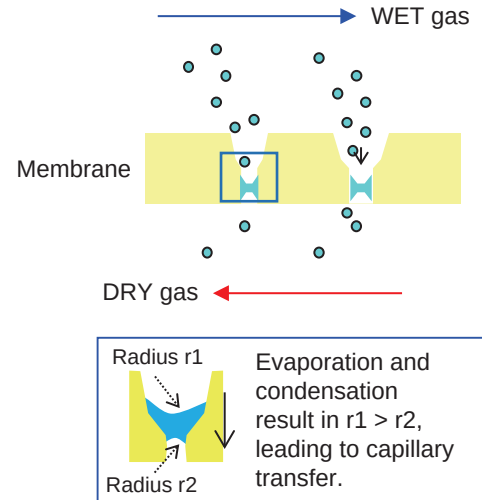


Fig. 5 Mechanism of water transfer

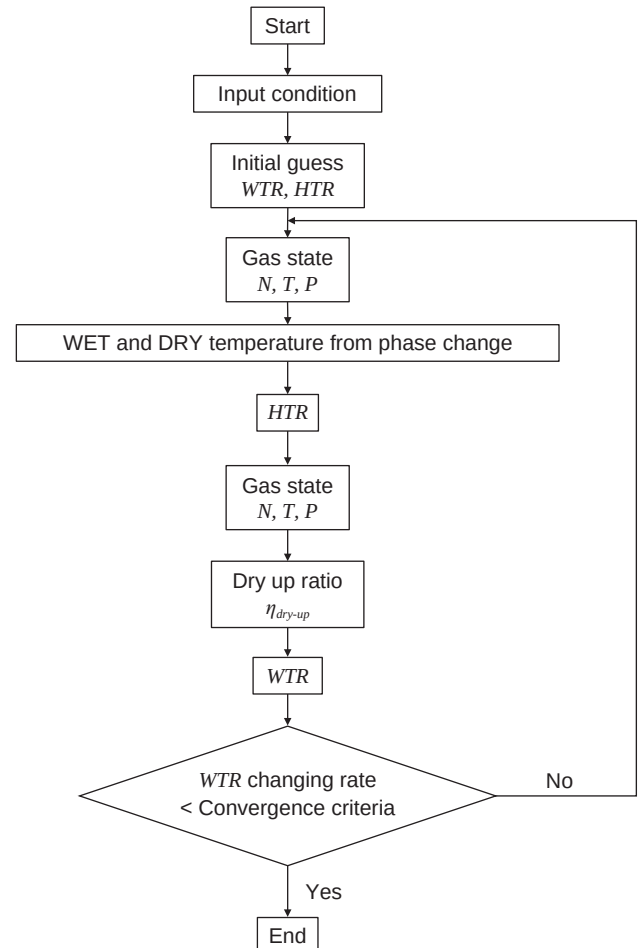


Fig. 6 Flowchart of humidifier model

The initial condition is a simple calculation of the water vapor and temperature distributions inside the humidifier, using the gas states at the inlet and outlet of the humidifier to be calculated as the inputs. Next, the gas temperature distribution is obtained from the heat transfer rate (HTR) between the WET gas and DRY gas and the latent heat of evaporation due to phase change between water vapor and liquid water, and the water transfer rate (WTR) is calculated based on the water vapor partial pressure and humidity in each part. In addition, due to the influence of the dry-up phenomenon, the dry-up ratio (η_{dry-up}) described in Section 3.4 is obtained from the gas temperature and humidity and taken into account in the WTR calculation.

The WTR of the membrane and the water vapor partial pressure distribution in the axial direction of the hollow fibers have a strong mutual influence. Therefore, accuracy was enhanced by discretizing and calculating in one dimension in the axial direction of the hollow fibers, as shown in Fig. 7. This was also done for the relationship between heat transfer and temperature.

3.3. Water Transfer

This section describes the method of modeling the phenomena inside the humidifier. Water transfer is a permeation phenomenon with the difference in water vapor partial pressure between the WET gas and DRY gas as the driving force. The water transfer rate N_{wtr} [mol/s] is expressed by Eq. (2) using the membrane surface area S [m²], the water vapor partial pressures in the WET gas $P_{v,wet}$ [Pa] and the DRY gas $P_{v,dry}$ [Pa], and the permeation resistance in bulk $R_{mt,total}$ [s/m].

$$N_{wtr} = \frac{S}{R_{mt,total}} \cdot \frac{P_{v,wet} - P_{v,dry}}{RT} \quad (2)$$

Based on the three-resistance model used to explain the process of mass transfer inside the membrane⁽⁸⁾, this permeation resistance is considered to be the sum of three resistances in series: (i) evaporation resistance, (ii) capillary transfer resistance, and (iii) adsorption resistance (Fig. 8). The evaporation resistance and adsorption resistance can be expressed as shown in Eq. (3) and Eq. (4) by the boundary layer equation and dimensional analysis⁽⁹⁾. The flow in the

vicinity of the hollow fibers has a Reynolds number of 1000 or less and can be regarded as laminar flow, and thus the laminar flow equation was adopted for the dimensionless number multiplier. The capillary transfer resistance is expressed here by Eq. (5) using the membrane-specific value k_m .

Evaporation resistance:

$$R_{mt,dry} = d_{mem-inner} / (\alpha_{dry} Re^{0.5} Sc^{1/3} D_{H_2O,Air}) \quad (3)$$

Adsorption resistance:

$$R_{mt,wet} = d_{wet,asm} / (\alpha_{wet} Re^{0.5} Sc^{1/3} D_{H_2O,Air}) \quad (4)$$

Capillary transfer resistance:

$$R_{mt,m} = 1/k_m \quad (5)$$

3.4. Dry-up Phenomenon

This section describes the details and modeling of the dry-up phenomenon.

Figure 9 shows the relationship between the WTR and the water content in the WET gas by comparing the values estimated by the model and the actual measurement results using the test apparatus described in Chapter 4. From Eq. (1) to Eq. (5), if the capillary transfer resistance is assumed to be constant, the WTR is nearly proportional to the water vapor partial pressure in the WET gas, and is expected to exhibit characteristics like the blue dashed line. However, the actual WTR was smaller than the expected transfer rate, as shown by the green dots. This indicates an increase in the capillary transfer resistance due to the dry-up phenomenon.

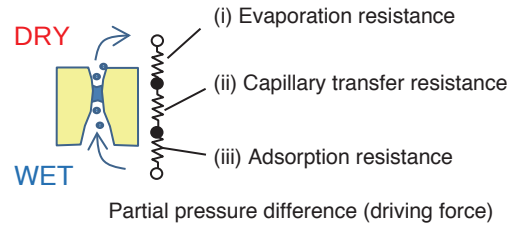


Fig. 8 Three-resistance model for water transfer

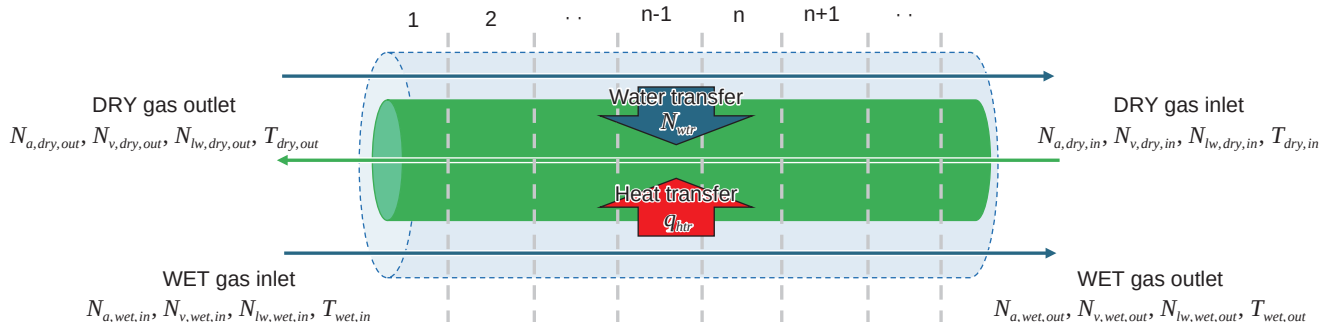


Fig. 7 Discretization of hollow fibers

Based on previous research on the mechanism of gas separation in porous membranes⁽¹⁰⁾, the following hypothesis was made about the mechanism of the drop in humidification performance due to the dry-up phenomenon. Figure 10 shows the mechanism of capillary transfer of water in porous material. The condensed water in the pores moves in the liquid phase with the gradient of the capillary attraction force produced by the difference in the Meniscus radius as the driving force, as shown in Fig. 10(a). When there is less water in the pores, the difference in the Meniscus radius becomes smaller, as shown in Fig. 10(b), and the transfer rate drops. Furthermore, evaporation produces vacancies on both sides inside the pores, and water vapor moves in these areas in the gas phase. Movement in the gas phase is Knudsen diffusion, in which the molecules move due to the pressure gradient while encountering resistance due to collision with the walls. However, the resistance is larger than that for movement in the liquid phase, such that resistance is added and the capillary transfer resistance increases. In pores with no condensate, movement is only by Knudsen diffusion, as shown in Fig. 10(c), and capillary transfer almost ceases to function.

The water vapor condenses inside the pores according to Kelvin's equation of condensation represented by Eq. (6)⁽¹¹⁾. When the RH of the gas inside the pores is outside the condensation range for the pore diameter and contact angle of the membrane, the condensate evaporates from inside the pores. Figure 11 shows the relationship between the RH of the WET gas and the pore diameter at the condensation limit. This shows that as the RH of the WET gas drops, the condensate evaporates from inside the pores at pore diameters larger than the pore diameter at the condensation limit. Figure 12 shows the pore diameter distribution per unit area of the porous membrane of the humidifier. Pores of various diameters are distributed in the membrane, and the condensate evaporates from an increasing number of pores as the RH of the WET gas drops and the pore diameter at the condensation limit becomes smaller. For pores in this state, the capillary transfer resistance increases as a result of less condensate, or the condensate disappears

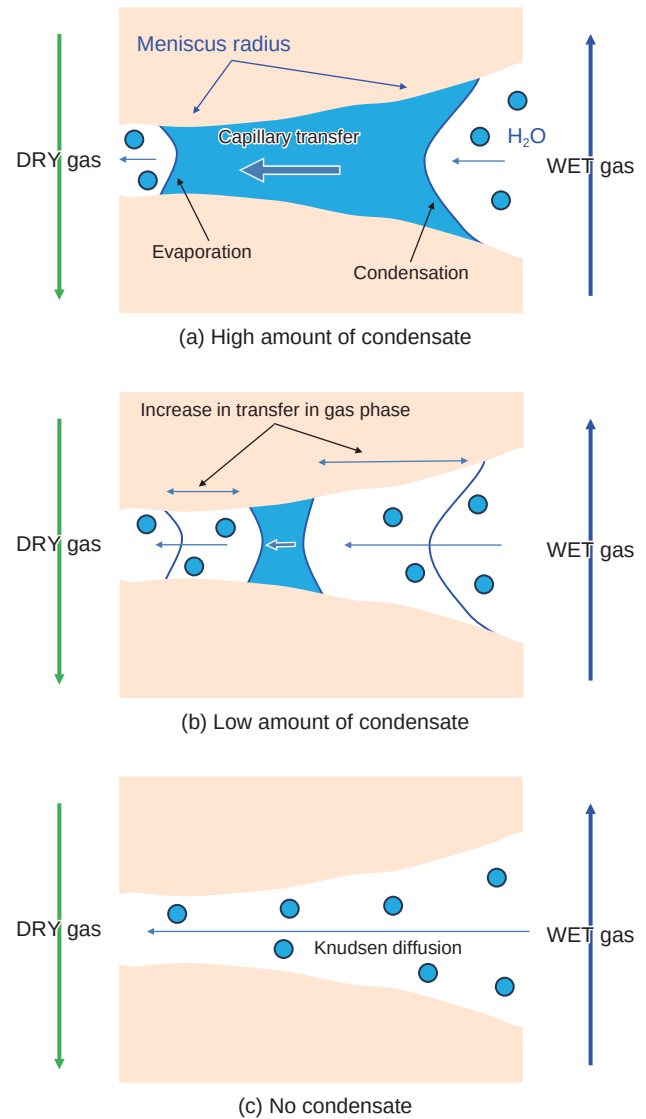


Fig. 10 Mechanism of capillary transfer

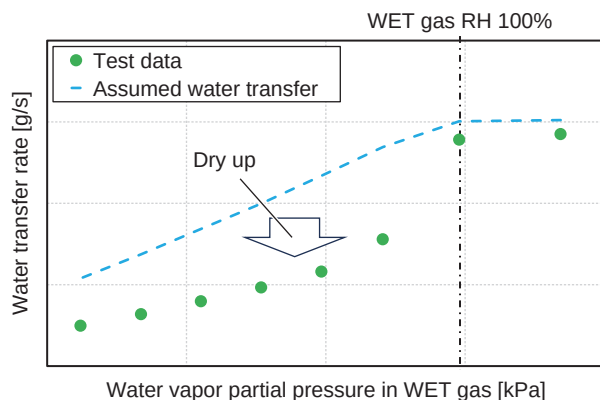


Fig. 9 Characteristics of dry up

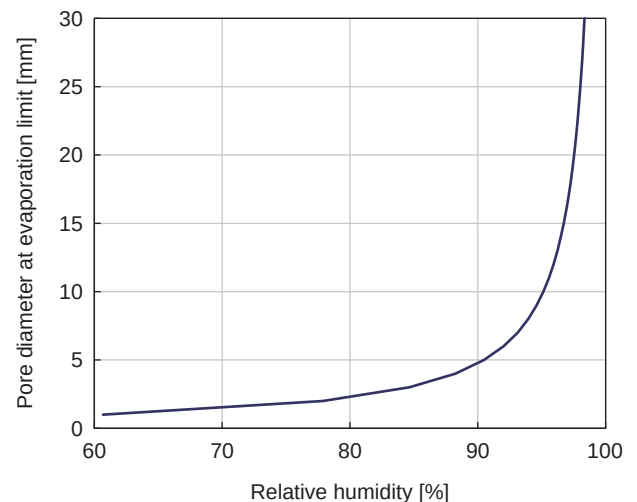


Fig. 11 Relationship between RH and pore diameter at condensation limit

and capillary transfer ceases to function, which is a factor resulting in lower humidification performance.

$$\ln \frac{RH}{100} = -\frac{2\gamma V}{rRT} \cos\theta \quad (6)$$

Here, the ratio of the number of pores at the condensation limit calculated from the Kelvin equation to the total number of pores in the porous membrane is defined as the dry-up ratio. In order to reproduce the drop in humidification performance due to the dry-up phenomenon in the model, the dry-up ratio is taken into account in the capillary transfer resistance in Eq. (5). It was observed with the test apparatus described in Chapter 4 that the capillary transfer resistance increases exponentially as the dry-up ratio increases, and thus a fitting parameter α_{dry-up} was set and this relationship was expressed by Eq. (7). α_{dry-up} uses the same value regardless of the operating conditions or humidifier specifications.

$$R_{mt,mem} = 1 / ((1 - \eta_{dry-up})^{\alpha_{dry-up}} k_m + \eta_{dry-up} k_K) \quad (7)$$

The capillary transfer resistance is influenced by the condensate in the pores, and thus a time lag was observed in the change in humidification characteristics due to the transition between the dry and wet conditions. Figure 13 shows the measured relationship between the humidifier weight and the WTR when the temperature of the DRY gas supplied to the humidifier is continuously varied to repeat the cycle of dry and wet conditions.

(A) is the state in which high-temperature DRY gas is flowed in, resulting in the dry condition. When the temperature of the DRY gas is lowered in this condition, the (B) wet condition results and the weight increases due to condensate. When the temperature of the DRY gas is raised to dry again, the (C) condition results in which the weight drops due to evaporation, but the WTR is about the

same as that in the wet condition. As shown here, hysteresis was confirmed in the relationship between weight change and the WTR, and the rate of drop in humidification performance due to evaporation and the rate of rise in humidification performance due to condensation were found to differ.

Figure 14 shows the drying process of condensate in the pores as the factor for this phenomenon. In this drying process, the evaporation of water is slower in the pores, and thus the condensate in areas that do not contribute to capillary transfer, such as the porous membrane surface, evaporates first. Therefore, it was thought that the capillary transfer resistance does not increase and the WTR is maintained until the condensate in the pores begins to evaporate.

A key to estimating the water transfer rate under transient conditions is to reproduce this hysteresis of change in the capillary transfer resistance. Therefore, in

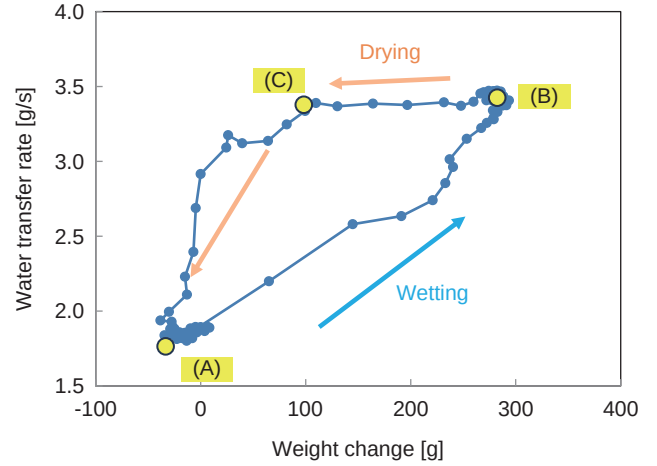


Fig. 13 Hysteresis of weight change versus water transfer

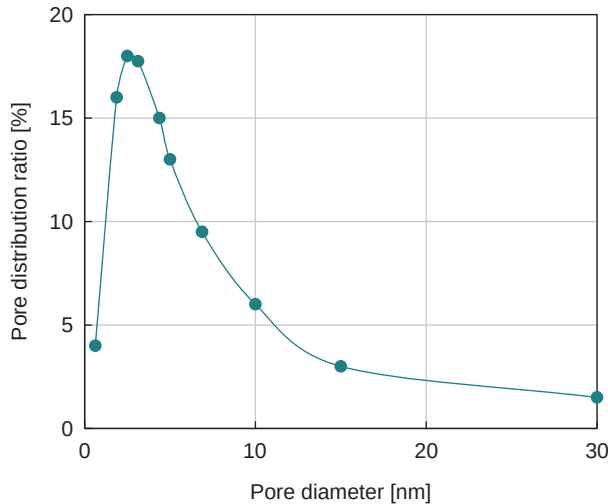


Fig. 12 Pore diameter distribution in membrane

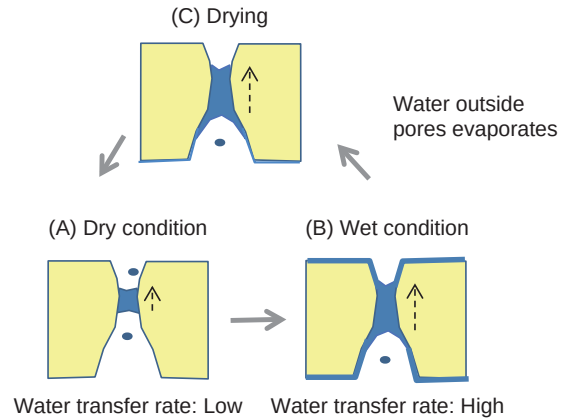


Fig. 14 Condensate during transition between dry and wet conditions

order to reproduce this time lag, the change in the dry-up ratio was defined by the first-order time-lag equations expressed by Eq. (8) and Eq. (9). As the condensation rate and evaporation rate have different characteristics, separate equations were used to enable separate coefficient settings.

Evaporation rate:

$$\frac{\partial \eta_{dry-up}}{\partial t} = \alpha_{eva} (\eta_{dry-up} - \eta_{dry-up}^{steady-state}) \quad (8)$$

Condensation rate:

$$\frac{\partial \eta_{dry-up}}{\partial t} = \alpha_{con} (\eta_{dry-up} - \eta_{dry-up}^{steady-state}) \quad (9)$$

3.5. Heat Transfer and Phase Change

Heat exchange between the WET gas and DRY gas is heat transfer with the temperature difference as the driving force. Like water transfer, it can be considered as series resistance, and is the sum of three resistances: heat transfer resistance from the WET gas to the WET side face of the membrane, heat transfer resistance from the WET side to the DRY side of the membrane, and heat transfer resistance from the DRY side face of the membrane to the DRY gas.

Furthermore, in heat transfer inside the humidifier module, the latent heat of evaporation influences the temperature distribution inside the module, and thus it is necessary to consider the evaporation of the condensate contained in the gas. The amount of evaporation N_{eva} [mol/s] can be expressed by Eq. (10) using dimensional analysis and boundary layer theory⁽¹²⁾. Therefore, the amount of heat generated was calculated from the amount of evaporation N_{eva} and the latent heat of evaporation.

$$N_{eva} = \beta D_{Air,H_2O} (2 + 0.37 Re^{0.61} Sc^{0.51}) \frac{P_{sat,v,wet} - P_{v,wet}}{RT} \quad (10)$$

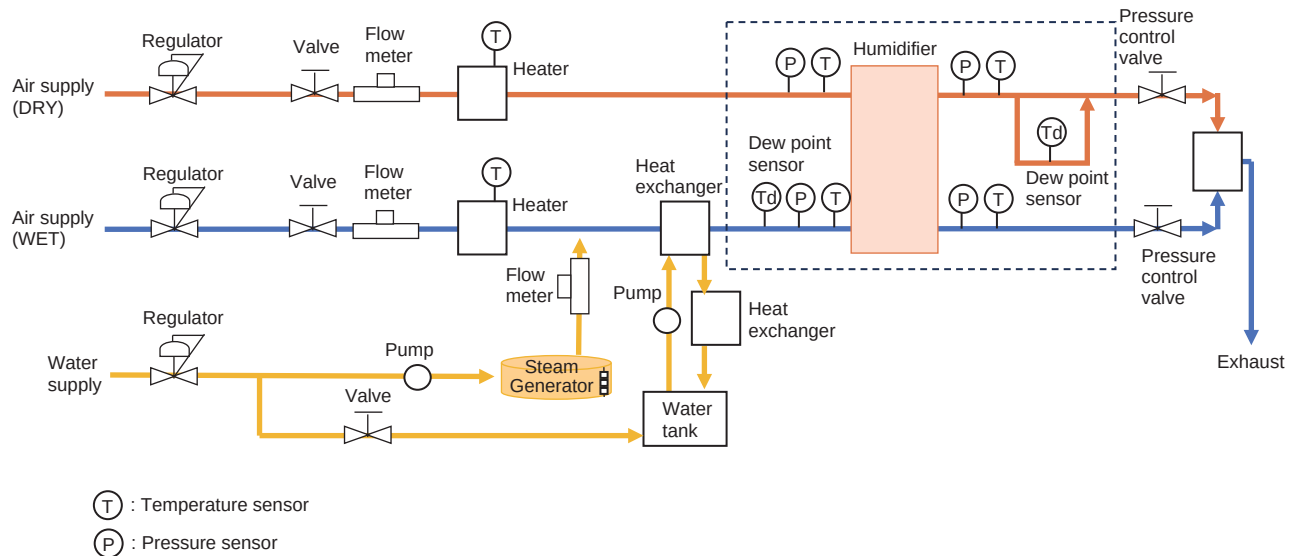


Fig. 15 Humidifier evaluation test apparatus

4. Verification of Humidifier Model Accuracy

The overall mechanism of humidification was modeled from the equations derived thus far, and a one-dimensional humidifier model was constructed. Chapter 4 compares the values estimated by this humidifier model and the actual WTR measurement data, and verifies the estimation accuracy of the model.

4.1. Verification of Model Accuracy by Humidifier Evaluation Tests

In order to verify the accuracy of the model under steady-state conditions, the model results were compared with the WTR measurement results obtained using a humidifier unit evaluation test apparatus. Figures 15 and 16 show the overall configuration of the humidifier test apparatus. In

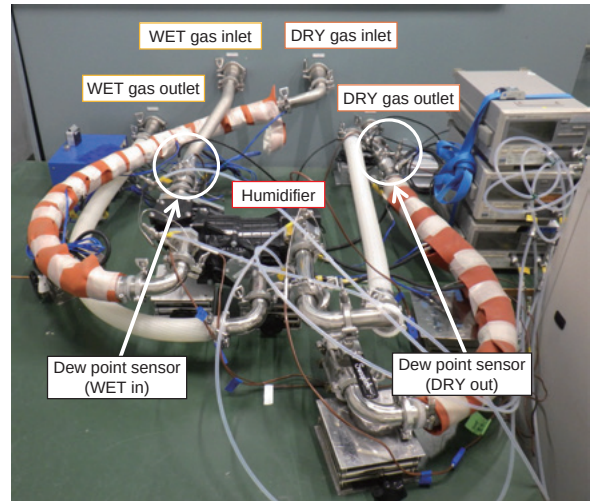


Fig. 16 Configuration around humidifier

Table 1 Gas conditions for humidifier evaluation test

Condition No.	Dry inlet temperature	Dry inlet pressure	Dry air flow rate	Dry water vapor flow rate	Wet inlet temperature	Wet inlet pressure	Wet air flow rate	Wet water vapor flow rate	Inlet wet ratio *
	K	kPaA	mol/s	mol/s	K	kPaA	mol/s	mol/s	%
1	333.2	237.0	2.87	0.000567	358.9	208.0	2.47	0.937	96.1
2	323.2	167.1	1.90	0.000533	351.2	140.0	1.66	0.712	96.2
3	313.7	145.7	1.52	0.000500	348.5	121.0	1.32	0.613	98.6
4	314.9	135.7	1.14	0.000400	349.7	116.0	0.97	0.547	101.7
5	315.4	125.8	0.76	0.000283	347.3	112.0	0.66	0.357	106.1
6	314.2	117.7	0.59	0.000233	340.7	107.0	0.50	0.190	105.7
7	313.4	112.0	0.38	0.000167	335.1	105.0	0.32	0.103	117.7
8	313.5	108.1	0.24	0.000100	332.0	103.4	0.20	0.060	126.7
9	313.2	105.5	0.14	0.000067	328.1	103.1	0.14	0.028	113.6

* Inlet wet ratio above 100% indicates presence of two-phase (liquid-gas) state. Inlet wet ratio up to 100% is equivalent to relative humidity (RH) .

Table 2 Gas conditions for humidifier evaluation test

Condition No.	Dry inlet temperature	Dry inlet pressure	Dry air flow rate	Dry water vapor flow rate	Wet inlet temperature	Wet inlet pressure	Wet air flow rate	Wet water vapor flow rate	Wet inlet water contents	Inlet wet ratio *
	K	kPaA	mol/s	mol/s	K	kPaA	mol/s	mol/s	g/s	%
10	338.4	224.0	1.756	0.00037	351.1	180.0	1.56	0.497	37.9	119.3
11								0.497	30.8	102.6
12								0.433	26.0	89.9
13								0.373	22.4	80.0
14								0.317	19.0	70.6

* Inlet wet ratio above 100% indicates presence of two-phase (liquid-gas) state. Inlet wet ratio up to 100% is equivalent to relative humidity (RH).

order to measure the WTR due to the humidifier, a method of flowing in the generated water vapor and mixing it into the dry air is adopted. The configuration enables realization of the predetermined test conditions by controlling the air flow rates with mass flow meters and adjusting the amount of water vapor generated to achieve the target humidity. Although this test apparatus is limited to steady-state conditions, evaluation can be performed under various operating conditions including low-humidity conditions. The accuracy of the model was verified by measuring the humidifier characteristics using this test apparatus and comparing them with the calculation results of the humidifier model under the same conditions.

Table 1 shows the supply air conditions assuming operating conditions from high load to low load in the fuel cell system. In the humidifier unit test, the WTR was measured while performing steady-state operation with the test apparatus set to these gas conditions. The gas conditions were set in the humidifier model as the input conditions, and the water transfer rate was calculated. Figure 17 compares the humidifier model and test results. As shown in the graph, it was confirmed that the humidifier model can estimate the WTR at an accuracy of $\pm 15\%$ or less under all conditions.

Next, the accuracy was also verified under conditions

where the dry-up phenomenon may occur. As shown in Table 2, tests and model calculations were performed even under gas conditions with a WET gas RH of less than 100%, and the results were compared. Conditions No. 12 to 14 are the region in which the dry-up phenomenon may

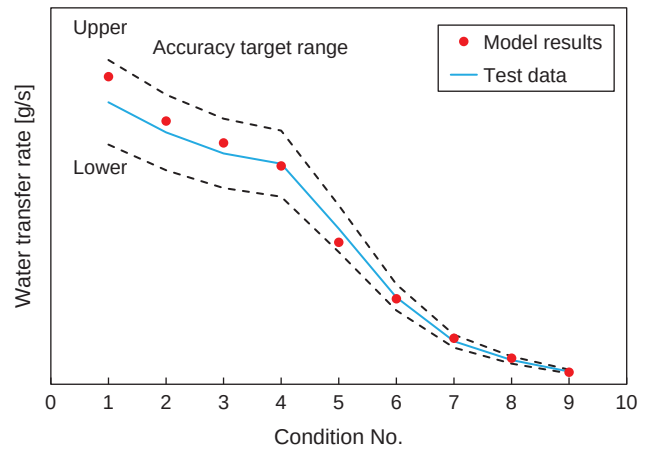


Fig. 17 Comparison of humidifier model and test results

occur. Figure 18 shows the comparison results. Here, the calculation results did not achieve the target accuracy when not considering the increase in the capillary transfer resistance. However, it was confirmed that the humidifier model that considers the dry-up phenomenon accurately reproduces the drop in WTR and can estimate the water transfer rate at the target accuracy of $\pm 15\%$ or less.

4.2. Verification of Model Accuracy in Fuel Cell System

As verification of the humidifier model accuracy under transient conditions, the model results were compared with measurement results of the RH of the supply air in fuel cell system operation. The fuel cell system performed power generation simulating the U.S. Environmental Protection Agency's (EPA) city driving mode, and the RH of the supply air was measured by attaching Vaisala Corporation's HUMICAP HMT337 dew point measurement instrument to the humidifier DRY gas outlet piping. Figure 19 compares

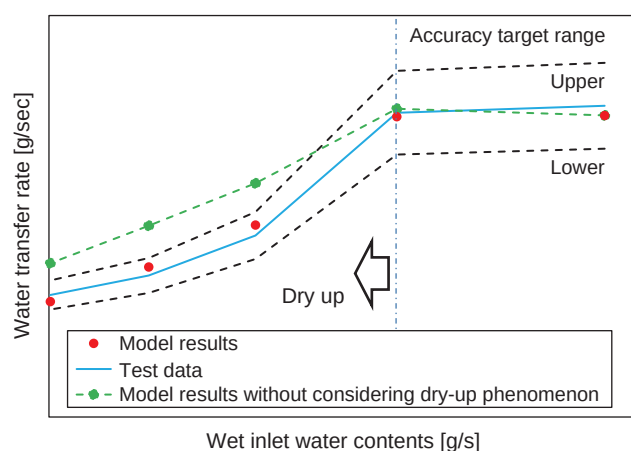


Fig. 18 Comparison of humidifier model and test results in dry up conditions

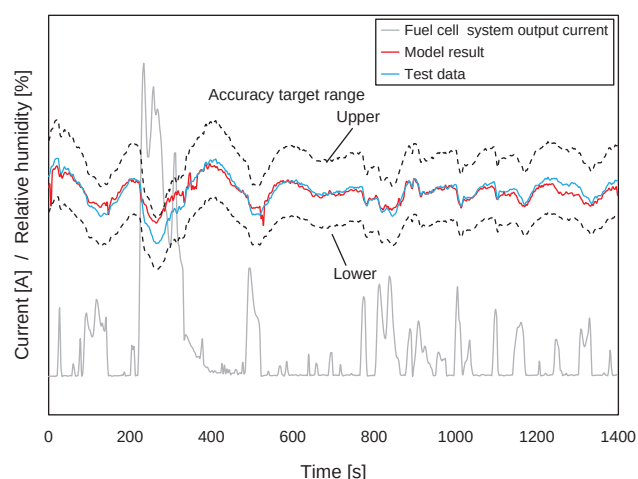


Fig. 19 Comparison of humidifier model and test results in mode operation

the time series data of the RH measurement results and the model calculation results. This confirmed that the model can estimate RH at the target accuracy of $\pm 15\%$ or less even under transient conditions.

In Honda's new fuel cell system, this one-dimensional humidifier model was converted to an approximation function, and a control model that provides accuracy while lowering the calculation load was implemented. Highly accurate water content control is achieved by controlling the coolant temperature based on the amount of water vapor calculated by this control model⁽¹³⁾.

5. Conclusion

A one-dimensional humidifier model that estimates the RH of the air supplied to the fuel cell stack from the operating conditions was constructed for use in performance design and control optimization of fuel cell systems.

- (1) The water transfer, heat transfer, and water phase change phenomena inside a porous hollow fiber membrane humidifier are expressed mathematically.
- (2) The drop in the WTR and the response time lag of that drop were modeled, focusing on the dry-up phenomenon.
- (3) The accuracy of the model was verified by comparing the model calculation results with the WTR measurement values in humidifier unit evaluation tests and measurement results of the RH of the supply air in bench mode operation of the fuel cell system. It was confirmed that the WTR due to the humidifier can be estimated at the set target accuracy of $\pm 15\%$ or less under both steady-state conditions including the dry-up phenomenon and transient conditions in the fuel cell system.

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List of Symbols

d	Pore diameter [m]
$D_{H_2O,Air}$	Diffusion coefficient between air and vapor [m^2/s]
k	Transfer coefficient [m/s]
N	Molar flow rate [mol/s]
P	Pressure [Pa]
q_{hr}	Heat transfer rate [J/s]
r	Radius of curvature of pore [m]
R	Gas constant [$J \cdot mol^{-1} \cdot K^{-1}$]
R_{mt}	Transfer resistance [s/m]
Re	Reynolds number
S	Membrane surface area [m^2]
Sc	Schmidt number
T	Temperature [K]
t	Time [s]
V	Molar volume of water [m^3/mol]
α	Fitting coefficient
β	Fitting coefficient
γ	Surface tension of water [N/m]
η_{dry-up}	Dry up rate
θ	Contact angle [degree]

Subscripts

a	Air
asm	Assumption
con	Condensation
dry	DRY side
$dry-up$	Dry up
eva	Evaporation
in	Inlet
K	Knudsen diffusion
l	Liquid water
m	Capillary transfer in membrane
$mem-inner$	Membrane inner
out	Outlet
sat	Saturation
$steady-stat$	Steady state
$total$	Total
v	Vapor
wet	WET side
wtr	Water transfer

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